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PROPERTIES AND EVOLUTION OF AEROSOLS WITH SIZE DISTRIBUTIONS HAVING IDENTICAL MOMENTS

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ABSTRACT

It has been proposed that the properties and evolution of an aerosol can be represented by lower order moments of its size distribution without requiring additional knowledge of the distribution itself. However certain distributions, including the log-normal and modified gamma distributions widely used to represent aerosol size distributions, belong to classes of multiple distributions having identical sets of moments, a situation that brings into question the utility of moment-based representations of aerosol properties and dynamics. We compare aerosol properties and evolution for explicit dissimilar test distributions having identical moments. It is found that despite their dissimilarity (e.g., multimode vs. single mode) these distributions exhibit virtually identical physical and optical properties and dynamics. This analysis allays the concern regarding applicability of moment-based representations of aerosol properties that arises out of the existence of sets of distributions exhibiting identical moments.

INTRODUCTION

Size distributions of aerosols are shaped by complicated nucleation, growth, and transport processes that are difficult to represent in models needed to describe the evolution of aerosols in multidimensional environments (e.g., models of atmospheric transport and turbulent jet flows). The size distribution is not only difficult to model numerically, it contains more information than is generally required. A possible alternative approach to explicit modeling of aerosol size distribution evolution in simulations of aerosol dynamics is that of tracking evolution of the aerosol in terms of the radial moments of the particle size distribution (this is the method of moments, MOM). The radial moments for a distribution of spherical particles are defined as:

$$\mu_k(t) = \int_0^\infty r^k f(r) dr \tag{1}$$

where $f(r)$ gives the fraction of particles in the radius range r to $r + dr$ and where k is the order of the moment. The moments are tracked either as a function of time, for spatially uniform systems (Friedlander, 1983; McGraw and Saunders, 1984; Pratsinis, 1988), or of space and time for simulations involving complex flows (Jurcik and Brock, 1993; LaViolette et al., 1996). In special cases, such as free-molecular growth, the equations governing the moment evolution are expressible in closed form in the low order moments of the distribution and yield exact analytical or numerical solutions for the spatial and temporal distributions of the moments. In other instances, accurate approximate solutions can be obtained by Gaussian quadrature (McGraw, 1997).

A collateral advantage of the MOM is that many physical and optical properties of aerosols (McGraw et al., 1997; Yue et al., 1997) and clouds (Hu and Stamnes, 1994) can be estimated directly from knowledge of the lower-order moments of the radial size distribution [or, equivalently, can be parameterized in terms of the effective radius which is itself defined as a ratio of lower-order moments (see Eq. 8 below)]. Thus moment parameterizations may be well suited for use in models describing the transport, evolution, and radiative properties of

atmospheric aerosols with key properties represented accurately in terms of low-order integral moments of the radial size distribution, without knowledge of the entire size distribution itself.

A possible concern with the above approach arises from the fact that certain size distributions are not uniquely determined by their integral moments, and *a fortiori* by their lower-order moments. In particular, this situation applies to the log-normal distribution for which, for any specified count median radius and geometric standard deviation, there exists an infinite set of related but dissimilar distributions having identical integral (both positive and negative) moments (White, 1990). The same situation is shown in the following section to apply for the modified gamma distribution and the non-negative integral moments. Given that the log-normal and modified gamma distributions are commonly employed as analytical representations of size distributions of atmospheric aerosols, this situation represents a potential problem for approaches attempting to characterize aerosols and represent their evolution in terms of the moments of their size distributions. Consequently, we address in this paper the extent to which the aerosol properties of distributions based on the log-normal and modified gamma distributions differ from those of the parent distributions. We examine the extent to which distributions having identical moments (which we refer to as isomomental distributions) exhibit different physical and optical properties and evolve differently in time. This study thus provides a stringent test of the ability of moment-based parameterizations to represent aerosol properties and evolution.

DISSIMILAR SIZE DISTRIBUTIONS HAVING IDENTICAL MOMENTS

Determining a positive distribution function from its complete set of non-negative integral moments is equivalent to solution of an integral equation of the first kind (Morse and Feshbach, 1953). Under certain conditions a unique distribution is obtained (Stoyanov, 1987). However, these conditions are not satisfied for certain distributions (Heyde, 1963; Stoyanov, 1987), and pertinent examples turn out to be distributions which have been widely employed as model distributions in aerosol science. This situation was noted by White (1990) who examined

properties of the log-normal distribution. Consider, for example, the normalized log-normal distribution:

$$f_{LN}(r) = (rs\sqrt{2\pi})^{-1} \exp[-(\ln r - m)^2 / 2s^2] \qquad (2a)$$

where r is the ratio of particle radius to the unit of length (typically 1 μm), m is the logarithm of the ratio of the count median radius (which is equal to the logarithm of the geometric mean radius) to the unit of length, and s is the logarithm of the geometric standard deviation (Hinds, 1982). With these definitions, $f_{LN}(r)dr$ gives the fraction of particles in the size range r to $r + dr$. The k th radial moment of Eq. 2a (e.g., Heintzenberg, 1994) is:

$$\mu_{LN}(k) \equiv \int_0^\infty r^k f_{LN}(r)dr = \exp[km + (ks)^2 /2] \, . \qquad (2b)$$

As shown by White, there is a set of distributions (denoted here Heyde distributions) based on the log-normal distribution,

$$g_{LN}(r) = f_{LN}(r)[1 + wF(\ln r - m)] \, , \qquad (2c)$$

where $F(x)$ is any odd function with period s^2 with $-1 \le F(x) \le 1$ and $-1 \le w \le 1$, having the same integral moments (positive, zero, and negative) as $f_{LN}(r)$; the conditions on w and F insure, among other properties, that $g_{LN}(r)$ is positive definite. Examples of these distributions where $F(x) = \sin(2\pi x / s^2)$ and $w = \pm 1$ are shown in the top panel of Fig. 1 for parameters such that the average particle size and the width of the distribution are typical of atmospheric aerosols (Whitby, 1978).

The modified gamma distribution (Pruppacher and Klett, 1980):

$$f_{MG}(r) = ar^n \exp(-br^s) \, , \qquad (3a)$$

where b is a constant and a normalizes the distribution, is also not determined uniquely by its moments. This can be most easily shown by extending the analysis of Stoyanov (1987). Stoyanov treats the case $n = 0$ and $0 < s < 1/2$, but his derivation (based on Eq. 4 below) holds also when n takes on other integer values; for example, the Nukiyama-Tanasawa particle size distribution (Fuchs, 1989) has $n = 2$. The moments of Eq. 3a are:

$$\mu_{MG}(k) \equiv \int_0^\infty r^k f_{MG}(r)dr = \frac{b^{-k/s}\Gamma[(k+n+1)/s]}{\Gamma[(n+1)/s]}. \tag{3b}$$

Now consider the distribution:

$$g_{MG}(r) = f_{MG}(r)[1 + w\sin(\beta r^s)] \tag{3c}$$

for $-1 \le w \le 1$ and $\beta = b\tan(s\pi)$. Inspection of Eqs. 3a and 3c reveals that provided

$$\int_0^\infty r^{k+n}\exp(-br^s)\sin(\beta r^s)dr = 0, \tag{4}$$

$g_{MG}(r)$ and $f_{MG}(r)$ will have identical moments. Equation 4 has been shown to hold for all non-negative integer values of $k+n$ and $0 < s < 1/2$ (Stoyanov, 1987). Thus these distributions have identical non-negative integral moments for $n = 0,1,2,...$ and $0 < s < 1/2$. As shown in the lower panel of Fig. 1, despite the fact that the $f_{MG}(r)$ and $g_{MG}(r)$ distributions have identical moments, they appear quite dissimilar.

PHYSICAL AND OPTICAL PROPERTIES

The existence of dissimilar distributions having identical moments requires reexamination of the utility of parameterizations of aerosol physical and optical properties based on moments. If significant differences in properties arise between different distributions having identical moments, the utility of moment based parameterizations of properties is called into question. Similar considerations apply to aerosol dynamics simulations based on the method of moments

(Friedlander, 1983; McGraw and Saunders, 1984; Pratsinis, 1988; Jurcik and Brock, 1993), which track the evolution of an aerosol via the evolution of the moments; to be useful it is necessary that the aerosol properties of interest be capable of reliable estimation from the tracked moments. In the following we show that despite the face that the moments do not uniquely define the distribution, moment parameterizations nonetheless give a good approximation to key aerosol properties.

We focus on single-particle derived properties of the aerosol, which can be expressed as an integral over the particle size distribution:

$$I = \int \sigma(r) f(r) dr \tag{5}$$

where $\sigma(r)$ is a kernel function that describes the physical property under investigation. In special cases $\sigma(r)$ is a simple power of r, or is polynomial in r, or can be expressed in polynomial form (McGraw et al., 1995). For these cases, property I is proportional to a moment or sum of moments, respectively, of the size distribution and will therefore be identical for isomomental distributions. Obvious examples of a power dependence include particle number, total surface area, and volume of the aerosol, which are proportional to the zeroth, second, and third moments, respectively. Likewise, the total surface area and mass fluxes of material sedimenting from a stationary fluid under Stokes flow, are proportional to the fourth and fifth moments, respectively, and Rayleigh scattering is proportional to the sixth moment (Friedlander, 1977).

Furthermore, limiting parameterizations to polynomial combinations of moments is unduly restrictive, and it is very likely that other moment combinations may be more effective. An important example is the parameterization of optical properties of aerosols and clouds in terms of the effective radius and effective variance (Hansen and Travis, 1974; Hu and Stamnes, 1994; Lacis and Mishchenko, 1995), defined as:

$$r_e = \frac{\int r^3 f(r) dr}{\int r^2 f(r) dr}. \tag{6a}$$

$$\nu_e = \frac{\int (r - r_e)^2 r^2 f(r) dr}{r_e^2 \int r^2 f(r) dr}. \tag{6b}$$

Although these quantities are not of polynomial form, it is clear that any physical or optical property that is capable of being parameterized in terms of r_e and ν_e must exhibit the same value for distributions having identical second, third, and fourth moments. Thus parameterization in terms of lower-order moments using different (i.e. non polynomial) functions of lower-order moments may yield substantially better results than indicated by the polynomial criterion. Retrieval of aerosol optical properties from moments has also been carried out through the intermediate step of obtaining estimates of the size distribution from the moments, followed by computation of optical properties from the estimated distributions (Yue et al., 1997). It is clear that all of these methods will yield identical predictions for isomomental distributions.

To be sure, there are cases where $\sigma(r)$ exhibits sufficiently complex behavior that I might not be simply expressed in terms of the moments of the size distribution. For example, the kernel function $\sigma(r)$ might represent a cross section for light scattering, as in Eq. 7 below, which will then exhibit the complicated structure dictated by Mie theory (Twitty, 1975). Nevertheless, despite the complexity of the Mie scattering kernel, even the lower-order radial moments of the aerosol size distribution have been shown to yield accurate parametrizations of the optical properties of aerosols (McGraw et al., 1995; Yue et al., 1997) and clouds (Hu and Stamnes, 1994). A stringent test of moment parameterizations is provided by predictions of the light scattering phase function for particles in the Mie size regime. The phase function specifies the angular distribution of scattered light and can be written as (Bohren and Huffman, 1983):

$$p_\lambda(\theta) = N \int S_{11}(r, \lambda, \theta) f(r) dr \tag{7}$$

where $S_{11}(r,\lambda,\theta)$ gives the angular distribution of scattered light from a single particle for unpolarized incident light, the angle θ is measured from the forward scattering direction, and N is a normalization constant. Figure 2 shows the phase functions for the distributions of Fig. 1 obtained using the computer codes of Hansen and Travis (1974). The phase functions for the several isomomental distributions are in excellent agreement even though the distributions themselves appear quite dissimilar in form. This agreement lends support to the supposition that the moments of the distribution can provide a reliable basis for predicting optical properties of particle distributions in the size range typical of atmospheric aerosols.

Extreme counterexamples include the case that $\sigma(r)$ is a delta function (or narrowly peaked function) centered on a specific radius r_0, which might be considered a functional representation of a mobility classifier. Inspection of Fig. 1 reveals that for such a function the integrals of Eq. 5 will not be identical for the different distributions (except of course at values of r_0 for which the distributions coincide). Similar considerations hold for the Heaviside function, which might be considered a model for an impactor or for the process of heterogeneous activation of an aerosol under supersaturated conditions.

AEROSOL DYNAMICS

The evolution of an aerosol size distribution can depend on a number of processes that include nucleation, hydrodynamic mixing, condensation/evaporation, coagulation, and mechanisms for particle loss. Moment methods, in which just the lower-order moments of the size distribution are tracked in space and time, can greatly simplify the representation of these processes in models (Friedlander, 1983; McGraw and Saunders, 1984; Pratsinis, 1988; Juric and Brock, 1993; McGraw, 1997). Any closed set of equations for evolution of the moments will, of course, be invariant to substitution of one isomomental distribution for another and therefore yield identical results. Of the processes governing evolution of the aerosol size distribution, those involving particle growth (e.g., condensation, evaporation, coagulation) introduce a dependence on the full size distribution and thus often limit closure of the dynamical equations governing moment evolution (McGraw, 1997). Under these conditions, even initially

isomomental distributions can be expected to evolve so as to have different sets of moments at later time. In this section we compare the manner in which different, initially isomomental, distributions evolve in time. To facilitate this comparison we consider only single-particle growth processes (and thereby exclude coagulation), for which analytical solutions are readily obtained (Clement, 1978). [Treatment of moment evolution under coagulation requires the introduction of more extensive numerical approximation schemes, some of which have recently been described (Barrett and Jheeta, 1996; Barrett and Webb, 1997).]

For an aerosol evolving according to the growth law $\varphi(r) = dr / dt$, the evolution of the k th moment can be represented in the general form described by Eq. 5 (Hulburt and Katz, 1964; McGraw, 1997):

$$\frac{d}{dt}\mu_k = k\int r^{k-1}\varphi(r)\,f(r)\,dr \tag{8}$$

for $k \geq 1$. For $k = 0$ we have $d\mu_0 / dt = 0$ as a consequence of the fact that although growth processes govern particle size, they do not change particle number. Equation 8 gives the contribution to the moment derivative due to particle growth. Other contributions to moment evolution, such as mixing of air parcels of different composition, will change particle number and introduce additional terms in Eq. 8 for the kth moment derivative. However, for a trace aerosol moving with the flow, these non-growth terms have a structure similar to those used to represent the mixing of trace chemical species in the same flow, generally do not affect closure, and can be handled by conventional fluid dynamics methods (LaViolette et al., 1996).

A commonly used expression for the rate of accretion of monomer by a particle during condensation growth under steady state conditions is:

$$I_T = \frac{b_M r^2}{r + a_M}. \tag{9}$$

This form also applies when heat transfer limits growth (Barrett and Clement, 1988). Considering only mass transfer, $b_M = 4\pi D(n - n_0)$ and $a_M = 4\lambda/3\alpha_M$ (Fuchs and Sutugin, 1971; Schwartz, 1986). In these equations, λ is the mean free path, α_M is the mass accommodation coefficient, D is the diffusion coefficient, n is the number density of condensable molecules in the vapor at large distance from the particle, and n_0 is the number density corresponding to the vapor pressure of the particle at equilibrium. Modified interpolation formulae have been presented, based on comparison with numerical simulation, that replace a_M with a size-dependent factor (Fuchs and Sutugin, 1971); however such expressions differ only slightly from Eq. 9 (Schwartz, 1986) and will not change the present results significantly, so we assume here that a_M is independent of particle size. Equation 9 reduces to the correct limiting expressions for particle growth in the free-molecular ($r << \lambda$) and continuum (diffusional) size limits ($r >> \lambda$). The growth law corresponding to Eq. 9 is

$$\varphi(r) \equiv \frac{dr}{dt} = \frac{v_1 b_M}{4\pi} \frac{1}{r + a_M} \tag{10}$$

where v_1 is the molecular volume of the monomer in the particle.

An analytic expression for the time evolution of a general aerosol size distribution under the growth law of Eq. 10, which conserves particle number, can be obtained from the continuity equation (Clement, 1978). The method furnishes the evolved aerosol distribution $f_t(r_t)$ at time t, for an arbitrary initial distribution $f_0(r)$ as follows: Conservation of particle number implies (Clement, 1978):

$$f_t(r_t)\,dr_t = f_0(r_0)\,dr_0 . \tag{11}$$

where r_0 is the initial particle radius and r_t is the particle radius after time t. Integrating the growth law of Eq. 10 gives

$$r_t^2 + 2a_M r_t = r_0^2 + 2a_M r_0 + \frac{v_1 b_M}{2\pi} t \tag{12}$$

Combining Eqs. 10-12 and expressing results in terms of the radius variable r_t, we obtain:

$$f_t(r_t) = \frac{r_t + a_M}{\sqrt{(r_t + a_M)^2 - \frac{v_1 b_M}{2\pi} t}} f_0\left[\sqrt{(r_t + a_M)^2 - \frac{v_1 b_M}{2\pi} t} - a_M\right] \tag{13}$$

where the prefactor of f_0 is in general dr_0 / dr_t. (Note that $f_t(r_t)$ is set to zero for negative argument of the square root.) The radial moments of $f_t(r_t)$ are obtained by substitution into Eq. 1 and carrying out the integrations numerically as described in the Appendix.

The growth law function $\varphi(r)$, appearing in the integrand of Eq. 8, is generally smoother and more amenable to polynomial approximation than is the light scattering kernel of Eq. 7. This suggests that isomomental distributions will tend to exhibit smaller differences in growth rate than in their light scattering properties. (In special cases such as free-molecular growth (Friedlander, 1977, 1983), where $\varphi(r)$ is independent of r, the integrand of Eq. 8 will be identical for all distributions that have the same moment sequence, and distributions that are initially isomomental will simply shift in the direction of increasing radius with time, thereby remaining isomomental throughout their evolution.) To illustrate the evolution of isomomental distributions for the growth law of Eq. 10, we choose, as the three initial distributions, the modified gamma parent distribution (3a) and the isomomental distributions (3c) for $w = \pm 1$. Size parameters have the same values as for the distributions shown in the lower panel of Fig. 1. Growth law parameters for the calculation were chosen such that $a_M = 4\lambda / 3\alpha_M = 1\,\mu\text{m}$ and $t_M \equiv 4\pi a_M^2 / v_1 b_M = 1\,\text{s}$. Thus for $\alpha_M = 1$, we set $\lambda = 0.75\,\mu\text{m}$. Setting the molecular volume equal to that of water ($v_1 = 3 \times 10^{-23}\text{cm}^3$) gives $b_M = 4.2 \times 10^{15}\text{cm}^{-1}\text{s}^{-1}$. In the Appendix we analyze moment evolution for a growth law having the general form of Eq. 10 and describe specific calculations for the parameters given above (Tables 1 and 2 and Fig. 4). As expected, deviations from the moments calculated for the parent distributions are indeed exceedingly small but nonzero (see Table 2 for numerical values).

It may be noted that the growth rate from Eq. 10, which is a decreasing function of particle radius, results in a narrowing of the particle size distribution as the distributions evolve with time. This is illustrated in Fig. 3, which shows the distributions at 5s, and Fig. 4, which shows the time dependence of the mean width and effective radius of the evolved distributions. The mean width of the distribution is obtained as:

$$\overline{W} = \left[\frac{\mu_2\mu_0 - \mu_1^2}{\mu_0}\right]^{1/2} . \qquad (14)$$

The effective radius is from Eq. 6. As a consequence of the near identity of the moments the mean widths and effective radii of the several distributions are virtually indistinguishable (see the Appendix for a more complete analysis of moment evolution).

DISCUSSION

In this paper we compared the physical and optical properties of aerosols having distributions characterized by the same sets of moments. Examples of differences in optical properties and in the time evolution of such isomomental distributions were evaluated. Because the full positive integral moment sequence is identical for these distributions, results from the present calculations provide a best-case limit (one in which all of the moments are available) on the accuracy of moment-based parameterizations of aerosol optical (Fig. 2) and growth (Figs. 3 and 4) properties. The moments of an aerosol size distribution have been shown to provide a reliable representation of physical and optical properties even when the size distributions themselves are not uniquely determined by their moments, as is the case for the distributions shown in Fig. 1. Additionally, there are prominent features of the size distribution evident in Fig. 1 which appear to be of secondary importance to the moments in determining physical and optical properties of the aerosol. For example, the three log-normal and three modified gamma distributions of Fig. 1 yield very similar phase functions (Fig. 2) even though in each case one of

the distributions is unimodal whereas the other two are distinctly multimodal and exhibit modes that are markedly displaced from each other.

Simulations of aerosol dynamics via the method of moments (Friedlander, 1983; McGraw and Saunders, 1984; Pratsinis, 1988; Jurcik and Brock, 1993; McGraw, 1997) track the moments of the size distribution directly in space and time. There are obvious advantages of combining such simulations, based on moments, with parameterizations for aerosol properties that are derived from the simulated moments. For example, one can imagine determining the radiative properties of atmospheric aerosols through regional-to-global scale simulations that track aerosol moments to investigate the role of aerosols in climate forcing. The moments of atmospheric aerosols can also be directly compared with properties inferred from ground based or satellite remote sensing measurements (Livingston and Russell, 1989).

Practical implementation of the method of moments would require using only a small number of the lower-order moments. If different distributions having identical moments were found to evolve in such a manner that their moments differed appreciably at later times, one would conclude that moments are not sufficient to parameterize the dynamical properties of the aerosol, even to the extent of predicting future values of the moments themselves. The results of numerical calculations reported here suggest that this is not the case. Furthermore, even the six lowest order moments provide an accurate parameterization for growth (McGraw, 1997) and optical properties (McGraw et al., 1995; Yue et al., 1997) of an aerosol. Nevertheless, as a note of caution, accuracy of moment-based parameterizations will depend both on the smoothness of the kernel function and the broadness of the particle size distribution - the ability to construct a low-order polynomial fit to the kernel function over the size range of the aerosol is a useful guide. Thus if indications are that more than six moments are required to obtain an accurate representation, a multimodal separation of the distribution, with separate moments for each mode, may be required. For example, if both unactivated aerosol particles and larger cloud droplets are present, it may be more reasonable (and more expedient) to use separate moments for representing and/or tracking of each of these clearly distinct particle types, than to treat all particles as part of a single distribution with a single set of moments.

SUMMARY AND CONCLUSIONS

Distribution functions can be constructed, based on the log-normal and modified gamma distributions, commonly used analytical representations of aerosol size distributions, that exhibit identical sequences of positive moments (and in the case of the log-normal distribution, negative moments also) despite substantial, qualitative differences--multimodal vs. monomodal and with significantly displaced modes. This situation calls into question the utility of representing aerosol properties and evolution in terms of the moments of their size distribution. We have therefore examined and compared light scattering phase functions and temporal evolution of aerosols having such isomomental size distributions. This examination shows that despite the considerable differences in the size distributions, the evolution and observable properties of such aerosols are in fact quite similar. This finding not only supports the utility of moment-based representations of aerosol size distributions and their evolution, but also suggests that for many applications the moment sequence may yield a more intrinsic representation of the aerosol than the size distribution itself.

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APPENDIX: Evolution of aerosol Radial MOMENTS ACCORDING TO THE GROWTH LAW OF EQ. 10

Evolution of the moments of a general aerosol size distribution

In this Appendix we use Eq. 8 and the interpolation growth law of Eq. 10 for numerical calculation of moment evolution. Without loss of generality we express r in units of $a_M = 4\lambda/3\alpha_M$ and time in units of $t_M = 4\pi a_M^2 / v_1 b_M$ yielding for the growth law of Eq. 10:

$$\tilde{\varphi}(\tilde{r}) \equiv \frac{d\tilde{r}}{d\tilde{t}} = \frac{1}{1+\tilde{r}}. \qquad \text{(A1)}$$

where the tilde indicates reduced units ($\tilde{r} = r/a_M$; $\tilde{t} = t/t_M$).

It is readily shown that for the growth law (A1) the time evolution of the reduced k th moment $\tilde{\mu}_k(\tilde{t})$ for any distribution is fully determined from the time evolution of $\tilde{\mu}_1(\tilde{t})$. This follows from the following identity:

$$\frac{\tilde{r}^{k-1}}{1+\tilde{r}} + \frac{\tilde{r}^{k}}{1+\tilde{r}} = \tilde{r}^{k-1}. \qquad \text{(A2)}$$

Combining Eqs. 8, A1, and A2 gives,

$$\frac{1}{k}\frac{d\tilde{\mu}_k}{d\tilde{t}} + \frac{1}{k+1}\frac{d\tilde{\mu}_{k+1}}{d\tilde{t}} = \tilde{\mu}_{k-1}. \qquad \text{(A3)}$$

Thus for any aerosol size distribution, and the growth law of Eq.A1, we obtain:

$$\frac{d\tilde{\mu}_2}{d\tilde{t}} = 2\tilde{\mu}_0 - 2\frac{d\tilde{\mu}_1}{d\tilde{t}}$$
$$\frac{d\tilde{\mu}_3}{d\tilde{t}} = 3\tilde{\mu}_1 - 3\tilde{\mu}_0 + 3\frac{d\tilde{\mu}_1}{d\tilde{t}} \qquad \text{(A4)}$$
$$\frac{d\tilde{\mu}_4}{d\tilde{t}} = 4\tilde{\mu}_2 - 4\tilde{\mu}_1 + 4\tilde{\mu}_0 - 4\frac{d\tilde{\mu}_1}{d\tilde{t}}$$
etc.

Evolution of the moments of the initially isomomental distributions

Consider the evolution of the radial moments for the three initially isomomental distributions, $w = 0, \pm 1$, based on the modified gamma distribution (Eq. 3c):

$$\mu_k(t) \equiv \int_0^\infty r^k g_{MG}(r)\, dr \qquad \text{(A5)}$$

where $g_{MG}(r)$ is evolved in time according to Eq. 13. The initially isomomental distributions lose that characteristic at later times. However the differences between the moments for the three distributions ($w = 0, \pm 1$) remain so extremely small that the calculation of these differences for the present study required high precision numerical integration (Wolfram, 1991).

An integral transformation based on the tangent function was used to handle the tail of the distributions at large r. Thus the integral of Eq. A5 was transformed using $z = \tan^{-1}(r)$ to obtain:

$$\mu_k(t) = \int_0^{\pi/2} [\tan(z)]^k g_{MG}[\tan(z)]\sec^2(z) dz \qquad \text{(A6)}$$

Evolved moments of the parent distribution at a sampling of time values are given in Table 1. The exceedingly small differences in the moments from the three evolved distributions are given in Table 2. Here we use the property that distributions add linearly in the moment integrals, and evaluate the moments of the difference of distributions as

$$\Delta\mu_k(t) \equiv \int_0^\infty r^k [g_{MG}(r) - f_{MG}(r)] dr \qquad \text{(A7)}$$

Table 1: Moments μ_k of the evolved modified gamma distribution (Eq. A5 for $w = 0$).
The units are μm^k.

t(s)	0	5	10	15	20
μ_1	0.11484	2.355	3.611	4.591	5.423
μ_2	0.03013	5.549	13.04	21.08	29.41
μ_3	0.01567	13.08	47.09	96.78	159.5
μ_4	0.01461	30.86	170.1	444.4	865.2
μ_5	0.02266	72.86	614.4	2040.5	4692.9

Table 2: Moments of the evolved difference distributions $\Delta\mu_k$ (Eq. A7 for $w = 1$).
The units are μm^k.

t(s)	0	5	10	15	20
$\Delta\mu_1$	0	3.03E-12	-3.64E-13	8.36E-15	1.38E-14
$\Delta\mu_2$	0	-6.06E-12	7.28E-13	-1.67E-14	-2.76E-14
$\Delta\mu_3$	0	1.47E-11	9.92E-14	-3.70E-13	-1.08E-13
$\Delta\mu_4$	0	-3.47E-11	-3.31E-12	1.55E-12	5.43E-13
$\Delta\mu_5$	0	-5.92E-11	1.92E-11	4.32E-12	-7 E-14

where $g_{MG}(r)$ is evaluated for $w = 1$, and $f_{MG}(r)$ is the parent distribution. Note that $\Delta\mu_k$ vanishes at $t = 0$ according to Eq. 4. The relative differences $\Delta\mu_k(t)/\mu_k(t)$ obtained from the ratio of corresponding entries of Tables 2 to those of Table 1, range from 1 part in 10^{12} to 1 part in 10^{17}. The numerical accuracy of the integration is confirmed by the result that $\Delta\mu_2 = -2\Delta\mu_1$, as required by the first of Eqs. A4, which from linearity, apply also to the moment differences (Note that since normalization is preserved, $\Delta\mu_0$ vanishes for all times).

The fact that the values of the differences $\Delta\mu_k(t)$ are exceedingly small, but nonzero (Table 1) is consistent with the polynomial criterion noted in the text. Nonvanishing differences imply that the growth function $1/(1+\tilde{r})$ does not have a convergent polynomial expansion. However the smallness of the nonisomomental tendency suggest that a good polynomial representation of the growth law can be obtained. The following observations on representation of the right-hand-side (rhs) of Eq. A1 by polynomials follow Lanczos (1988): The geometric expansion:

$$\frac{1}{1+\tilde{r}} = 1 - \tilde{r} + \tilde{r}^2 - \tilde{r}^3 + ... \qquad \text{(A8)}$$

has a convergence limit of unity and therefore the rhs of Eq. 1 does not have a convergent expansion over the radius interval $(0, \infty)$ of the integrated aerosol distributions. However a superior polynomial representation is available (one with greater accuracy and extended radius of convergence over a finite interval $0 \le \tilde{r} \le L$) based expansion of $1/(1+\tilde{r})$ in shifted Chebyshev polynomials ($T_n(2\tilde{r} - 1)$). This expansion has the form (Lanczos, 1988):

$$\frac{1}{1+\tilde{r}} = \sqrt{2}\{(1/2) - d \times T_1(2\tilde{r} - 1) + d^2 \times T_2(2\tilde{r} - 1) - d^3 \times T_3(2\tilde{r} - 1) + ...\} \qquad \text{(A9)}$$

where $d = 3 - 2\sqrt{2} = 0.1716...$. Comparisons for the same polynomial order (note that $T_n(2\tilde{r} - 1)$ is a polynomial of order n) show that Eq. A9 yields a much better polynomial representation of the growth law than is provided by the simple geometric expansion of Eq. A8.

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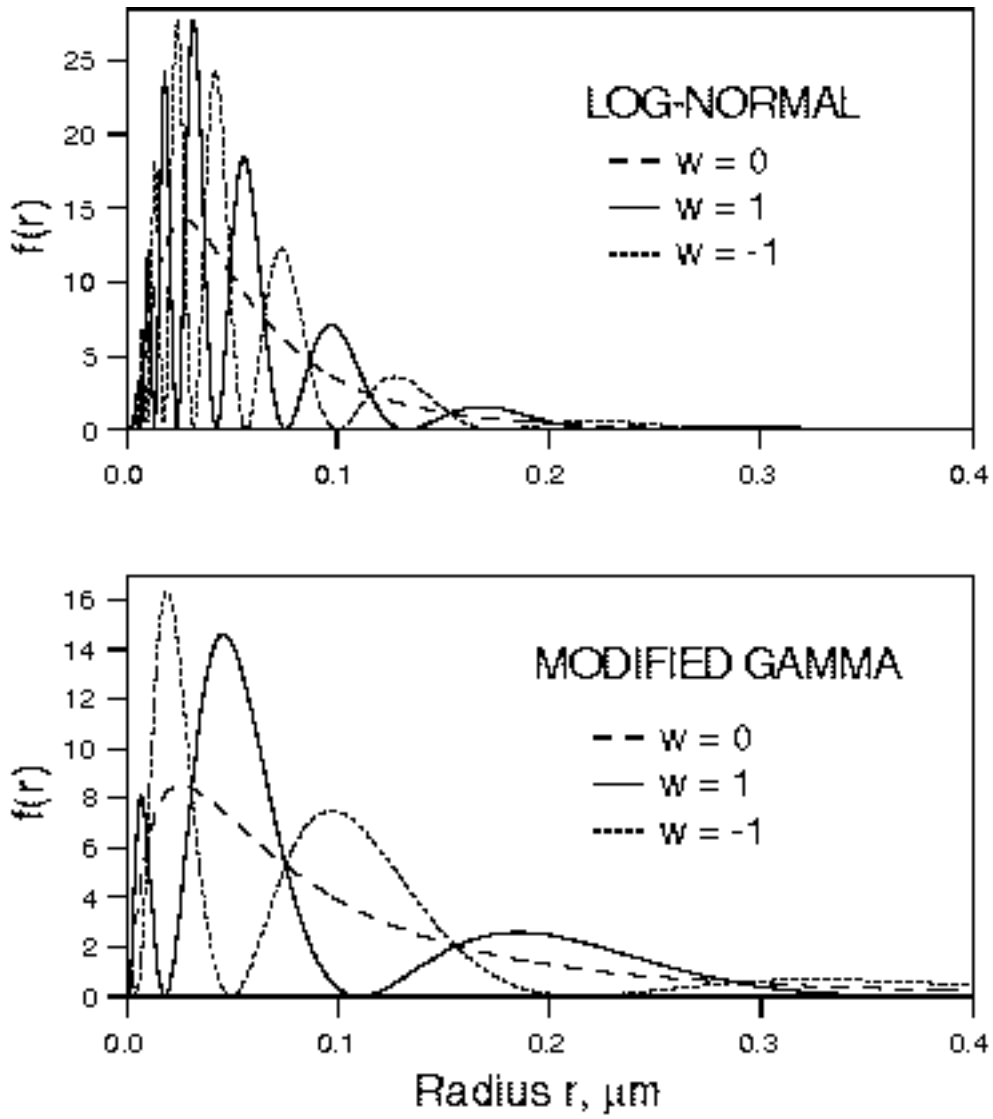

Figure 1. Two classes of size distributions having identical radial moments. Upper panel: dashed curve, log-normal distribution (Eq. 2a) for s=0.75 and m=-3; multimodal curves are the corresponding Heyde distributions from Eq. 2c for $F(x)=\sin(2\pi x/s^2)$. Lower panel: dashed curve, normalized modified gamma distribution (Eq. 3a) for n=3, b=30 and s=1/4; for these parameters the normalization constant $a = sb^{(n+1)/s} / \Gamma[(n+1)/s] = 8.23\text{x}10^{10}\mu\text{m}^{-4}$; multimodal curves are the corresponding Heyde distributions from Eq. 3c.

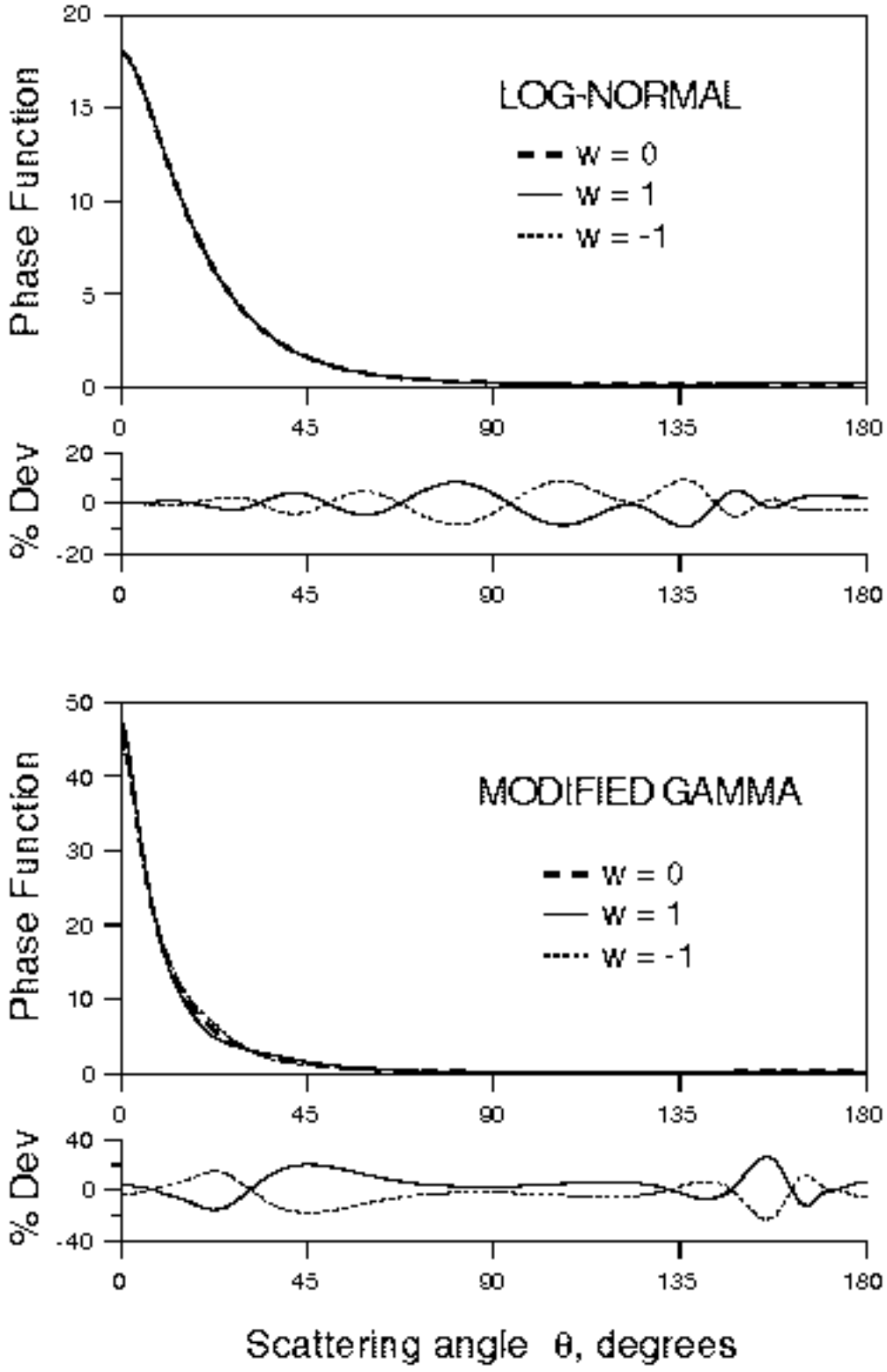

Figure 2. Phase functions for the distributions shown in Fig. 1 at a wavelength of 550 nm. Normalization for the phase functions has been chosen such that the integral over all scattering directions is 4π. The real part of the particle refractive index was set at 1.4, independent of particle size, and the imaginary part set to zero (no absorption). The percent deviation of the phase functions for the Heyde distributions from those for the corresponding parent distributions is also shown.

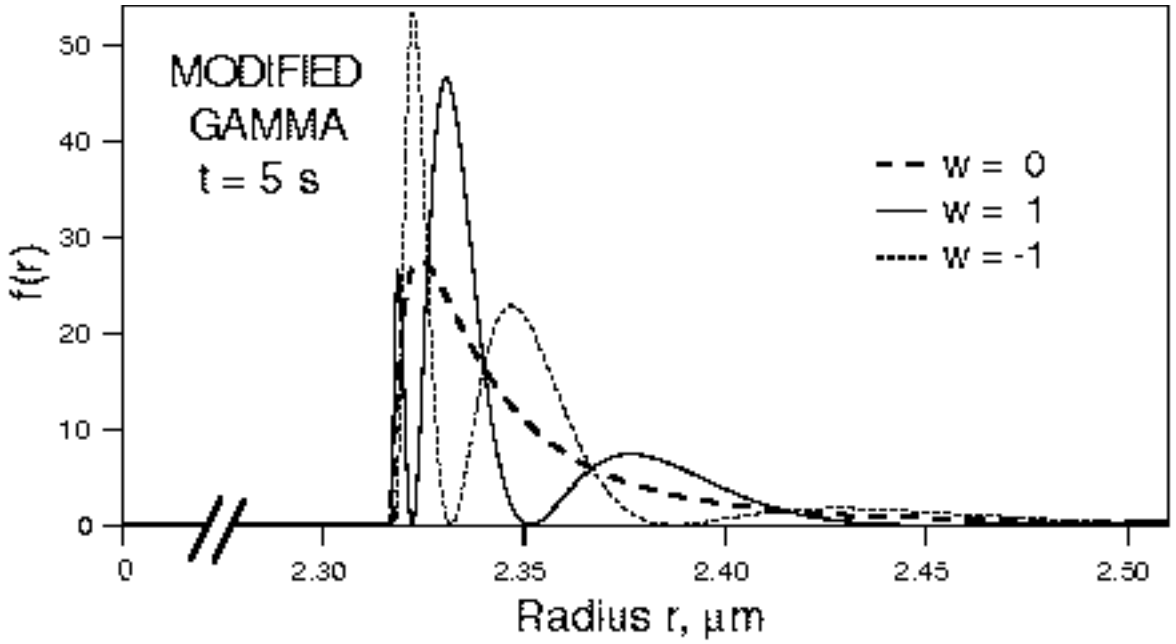

Figure 3. Distribution profiles at t=5s (from Eq. B4). Dashed curve, evolved normalized modified gamma distribution (Eq. 3a and Fig. 1 give the distribution at t=0) for n=3, b=30 and s=0.25; multimodal curves are the corresponding evolved Heyde distributions (Eq. 3c and Fig. 1 give the distributions at t=0).

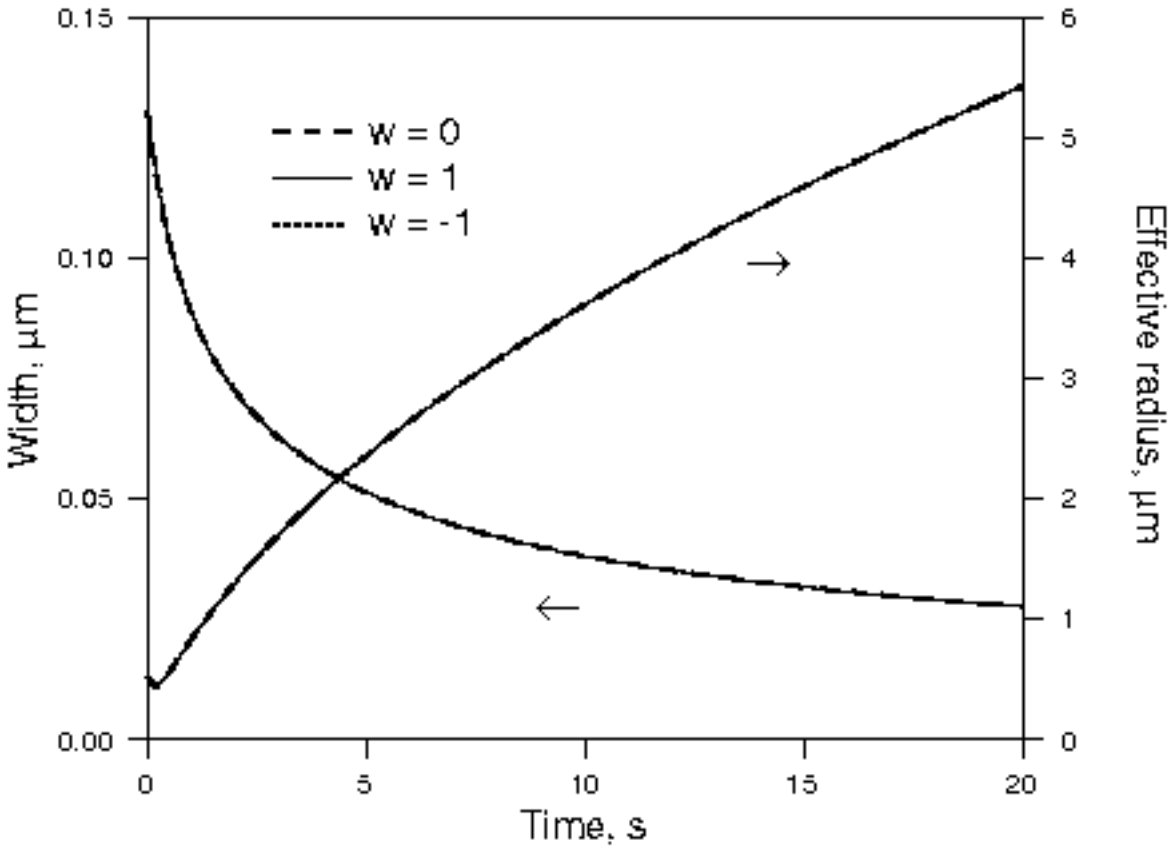

Figure 4. Time dependence of the widths of the evolved MG and derived Heyde distributions. The distribution widths were computed from the moments using Eq. 15. The figure also shows evolution of the effective radius according to Eq. 8 (right axis).